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(54) High pressure reactor for the synthesis of melamine

(57) Reactor for the synthesis of melamine from urea, in accordance with the highpressure non-catalytic process, comprising: a vertical reactor body (1), at least one inlet (2) for the urea melt, a set of heating elements (3), and a central duct (7), said set of heating elements (3) being arranged inside said central duct.

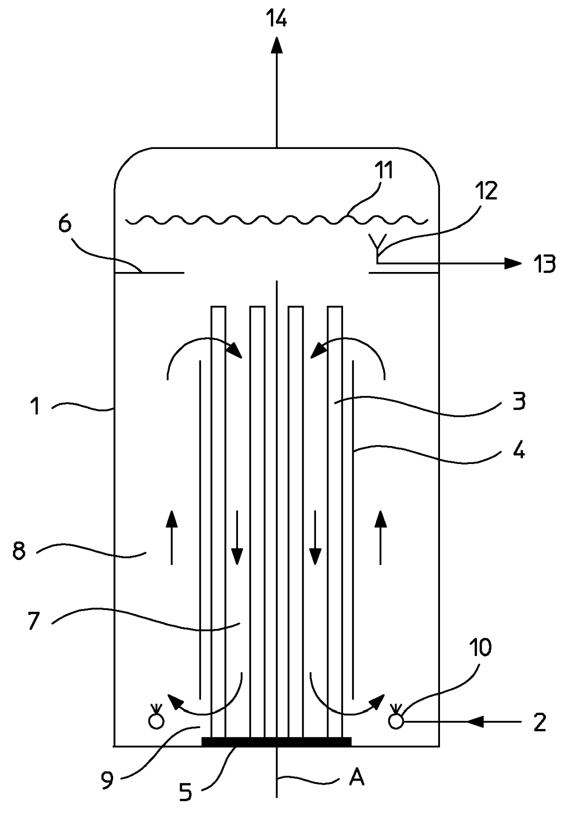

Fig.1

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Description

Field of application

[0001] The invention relates to a high-pressure reactor for the synthesis of melamine.

Prior art

[0002] The processes for the synthesis of melamine from urea are commonly classified as low-pressure catalytic processes, typically below 1 MPa, and high-pressure non-catalytic catalytic processes, typically above 7 MPa. These processes are well-known in literature (see for example Ullmann's Encyclopedia of Industrial Chemistry, 6th ed., vol. 21, p. 205).

[0003] One of the known high-pressure synthesis processes, as described for example in US 6,815,545, comprises essentially three steps: an endothermic reaction which converts urea into melamine inside a first reactor, called primary reactor; a second step for removal of the carbon dioxide (CO2) by introducing gaseous ammonia and for reduction of the content of by-products, which are converted into melamine with increase in the conversion, inside a second reactor called secondary reactor or stripping reactor; a third step during which the gases separated in the top part of the primary and secondary reactors (called "off-gases") are conveyed away for washing or scrubbing with urea before being conveyed to the urea plant.

[0004] A lay-out of a plant which operates using this process is typically the following.

[0005] The plant comprises a primary reactor, a secondary reactor and a scrubber, which are formed as separate cylindrical bodies. The urea melt is supplied to the primary reactor where the first reaction stage takes place that is the endothermic conversion into melamine; the effluent of said reactor is directed to the second reactor, where it undergoes a process of stripping of the gases contained therein, by means of gaseous ammonia. In general, the liquid melamine is kept inside this secondary reactor for a certain dwell time (aging of the melamine) in order to convert the by-products formed inside the primary reactor into melamine. The liquid effluent from the secondary reactor (melamine melt) may be sent to a subsequent further purification step.

[0006] The gases released inside the primary reactor and inside the secondary reactor form a stream of so-called off-gases, mainly containing ammonia and CO2 with small amounts of melamine; said stream of off-gases undergoes washing with urea melt inside the scrubber. The urea melt is thus heated before being supplied to the primary reactor; the off-gases at the scrubber outlet, which are melamine-free, are conveyed away and for example recycled for urea synthesis.

[0007] The pressure is generally between 70 and 250 bar (7-25 MPa) and typically about 100-120 bar (10-12 MPa).

[0008] US 6,815,545 describes a primary reactor according to the prior art, which essentially comprises: a vertical cylindrical body; a central duct; a bundle of heating tubes arranged in a ring around said central duct. The urea supply is introduced at the bottom of the central duct. According to the prior art, this reactor works substantially with a circulating flow which ascends inside the central duct and descends in the annular region occupied by the heating tubes.

[0009] Said primary reactor design is widely used in the prior art, but has a number of drawbacks.

[0010] The tube bundle has an outer diameter which is almost equivalent to the diameter of the reactor body, i.e. the entire apparatus. This means that, since it is required to be able to remove the tube bundle for periodic maintenance, a fully open flange with a tubesheet which has substantially the same diameter of the reactor - and is therefore thick, heavy and costly - is required. It should be noted that the reactor operates at high pressure (typically above 100 bar) and consequently the large-size flanges and the openings have a very high cost and may create problems as regards tightness.

[0011] Another drawback consists in the fact that the heating tube bundle is difficult to construct and hence costly; in particular, the heating tubes are bayonet (double-wall) tubes and require a double circular-rim tube plate, one of which has a large thickness.

[0012] A third drawback is that the urea inlet in the centre of the heating bundle must be disconnected/connected in every operation. Said inlet, however, is difficult to access and increases the downtime and the risk of leakages from the seals owing to the difficult accessibility of the respective flanged joint.

Summary of the invention

[0013] The invention aims to overcome the drawbacks and problems mentioned above.

[0014] This purpose is achieved with a reactor and a process for the synthesis of melamine from urea using the high-pressure non-catalytic process, according to the accompanying claims.

[0015] The reactor comprises a vertical body, at least one inlet for the urea melt, a set of heating elements, and a central duct, and is characterized in that said set of heating elements is arranged inside said central duct.

[0016] Said central duct is advantageously delimited by a shell, preferably cylindrical, which surrounds the set of heating elements. This shell may be defined as being a low-pressure shell situated inside the reactor. The outer shell of the reactor may be defined as being a high-pressure shell, since it withstands the high pressure of the process.

[0017] The central duct delimits an inner reaction zone and a peripheral reaction zone which communicate with each other and inside which a circulation flow is formed. The peripheral zone has the form of a circular rim and may be externally delimited by the shell of the reactor itself (high-pressure shell) or possibly by another inner shell. Advantageously, according to the invention, said peripheral zone around the central duct does not have any heating elements.

[0018] According to the invention, the heating elements are arranged in the centre of the reactor, i.e. in the inner zone, rather than in the peripheral zone around the duct. Preferably, the set of heating elements is a bundle of tubes which are connected to a tube plate at the bottom of the reactor and supplied with a suitable thermal fluid.

[0019] In a preferred embodiment, the reactor comprises a distributor for the urea supply, which is connected to said urea melt inlet and is configured to introduce urea into said zone. More advantageously, said distributor is configured to introduce urea in a substantially uniform manner and with axially symmetry into said peripheral zone.

[0020] In a preferred embodiment, said distributor is a toroidal body with a plurality of holes for introducing urea, which are distributed along its circumference; the distributor holes are directed so as to favour mixing of the urea melt with the fluid exiting the heating bundle, at the base of the central duct.

[0021] In an even more preferred embodiment, said central duct extends at a predefined distance from a base of the set of heating elements, thus defining a passage for the recirculation of liquid. Even more advantageously, said central duct extends, inside the reactor, up to a height lower than the height of the heating elements.

[0022] As a result of the configuration described it is possible to create a descending flow inside the central duct in the presence of the heating elements and an ascending recirculating flow in the surrounding annular zone (or rim), where there are no heating elements. Consequently, a descending flow is obtained in the heated reaction zone, as in the reactors of the prior art. This constitutes an advantage because the process conditions which are familiar to persons skilled in the art are retained, but with the constructional advantages arising from positioning the heating tubes in the central part of the reactor.

[0023] The mixing of the two fluids is ensured by the axial symmetry inside the annular cavity.

[0024] A deflector in the form of a circular rim is preferably installed at the top end of the peripheral reaction zone. Said deflector prevents turbulence on the free surface of the fluid inside the reactor and has the function of directing the flow into the inner zone containing the heating bundle. In certain preferred embodiments the header for the liquid melamine may be associated with said deflector.

[0025] The invention also relates to a process for the high-pressure synthesis of melamine from urea, comprising a primary reaction stage in which a stream of urea is converted into melamine with an endothermic reaction, inside a vertical reactor, characterized in that:

said primary reaction stage is performed with a flow circulating inside a liquid mass comprising melamine and urea, comprising a descending flow in a substantially column-like region in the centre of said reactor, directly heated by the presence of heating elements, and an ascending flow in a substantially rim-like region around said column, without heating elements, inside which the urea supply is introduced.

[0026] The invention has the following advantages. The set of heating elements is less complex, more compact, easier to manufacture and therefore less costly than the conventional reactors, owing to the positioning in the centre of the reactor. In fact, the heating elements are arranged in a substantially cylindrical zone in the centre of the reactor, instead of in an annular zone with the same outer diameter of the reactor itself.

[0027] With reference, for example, to heating elements in the form of tubes, the tubesheet has a smaller diameter and a smaller thickness, and the reactor body has a lower cost too because the flange coupling with the tubesheet is partially open rather than fully open. Owing to the proposed configuration, the periodic operations of disassembly of the heating bundle are simplified since it is no longer required to remove and restore the connections of the urea melt supply line. Consequently the plant downtimes are reduced. Finally it is possible to design and construct large-size reactors more easily owing to the greater simplicity and the smaller dimensions of the heating bundle.

[0028] In some embodiments, a reactor according to the invention may also incorporate a second reaction chamber, also called secondary section for stripping the liquid melamine with ammonia, and/or a section for scrubbing the gases.

[0029] The detailed description which follows relates to a preferred embodiment, described by way of a non-limiting example.

<u>Description of the figures</u>

[0030]

Fig. 1 is a schematic cross-section of a primary reactor for the high-pressure synthesis of melamine, according to a preferred embodiment of the invention.

Fig. 2 shows a variant of the reactor shown in Fig. 1, according to another embodiment of the invention.

<u>Detailed description</u>

[0031] Fig. 1 shows an example of a reactor which comprises a vertical body 1, an inlet 2 for the urea melt and a bundle of heating tubes 3 inside a central duct 7, delimited by a cylindrical shell 4.

[0032] The tubes 3 are fixed to a tubesheet 5 which is located at the bottom of the reactor.

[0033] The body 1 and the shell 4 are substantially axially symmetrical; preferably both the body 1 and the shell 4 are cylindrical.

[0034] Said shell 4 may be defined as being a low-pressure inner shell. In fact, it remains immersed inside the liquid melamine, during the functioning of the reactor, and is not subject to a substantial difference in pressure.

[0035] The shell 4 extends vertically inside the reactor up to a height which is advantageously lower than the height of the tubes 3, as shown.

[0036] The shell also delimits a substantially annular shaped region 8 outside the duct 7. Said region 8 forms a peripheral reaction zone around the central duct 7. In the example according to Fig. 1 said region 8 is delimited between the shell 4 and the cylindrical body 1; in other embodiments, however, the outer wall of said region 8 may be delimited by another low-pressure shell inside the body 1.

[0037] The bottom edge of the shell 4 is spaced from the tube plate 5, leaving a passage 9 for recirculation of the liquid.

[0038] The urea supply line 2 is connected to a toroidal distributor 10 provided with a plurality of distribution holes along its circumference. Thus configured, the distributor 10 introduces urea in a uniform manner into the annular region 8.

[0039] Advantageously said urea distributor 10 is at the base of the duct 7, as shown in Fig. 1, in the same region of said recirculation passage 9. In some variants (not shown) the toroidal body of said distributor 10 may be positioned on the outer diameter of the reactor body 1 or outside the reactor itself, so as to be accessible externally.

[0040] A diaphragm 6 is advantageously provided at the top of the annular region 8.

[0041] Under normal operating conditions, the reactor is almost completely full of liquid, as indicated by the level 11. The flow line 13 indicates the crude melamine exiting via a suitable header 12. The flow line 14 indicates the gases mainly containing ammonia and CO2 (off-gases) which are extracted from the top of the reactor.

[0042] The arrows in Fig. 1 indicate the axially symmetrical recirculating flow which is formed inside the reactor. Inside the central duct 7 a descending flow is generated, said flow entering the base of the annular section 8 via the passage 9, and being mixed with the urea supply. Inside the annular section 8 an ascending flow is created, assisted by the formation of bubbles in the liquid phase. Part of the liquid mass which emerges from the top of the annular section 8, also as a result of the deflector 6, recirculates with a descending flow and enters again the duct 7 via the open top end of the shell 4.

[0043] The conversion of urea into melamine takes place in the zones 7 and 8 in accordance with the known reaction: 6 urea $\rightarrow$ melamine + 6 NH_3 + 3 CO_2 (off-gas).

[0044] From Fig. 1 it is possible to better appreciate a number of advantages of the invention and in particular: the flange 5 has relatively small dimensions, owing to the central arrangement of the tube bundle 3; the circulating flow inside the reactor descends inside the region directly in contact with the heating elements (i.e. inside the duct 7) and ascends inside the annular portion 8.

[0045] The reactor has a substantially radial symmetry with respect to the axis A. In particular, the duct 7, the annular chamber 8 and the distributor 10 have a substantial radial symmetry with respect to said axis A. Therefore, the reactor may be defined as being axially symmetrical and the flow of the liquid is substantially axially symmetrical too.

[0046] Fig. 1 shows an embodiment in which the melamine 13 is drawn off at a specific point via the header 12.

[0047] In the variant according to Fig. 2, the header for collecting the crude melamine is advantageously formed above the deflector 6, thus allowing the product to be uniformly collected along the whole circumference of the reactor. More particularly, in this embodiment the reactor comprises a melamine header 15 having a top peripheral edge 16 positioned above said deflector 6. The melamine emerges through said header 15 and, once reached the edge 16, it flows out onto the deflector 6 which acts as an element for collecting the melamine. In this embodiment, the collection of melamine takes place in a distributed and substantially axially symmetrical manner along a circumference formed, in the example, by the edge 16. The advantage of an improved symmetry of the flows is thus obtained.

Claims

1. Reactor for the synthesis of melamine from urea, according to the high-pressure non-catalytic process, comprising: a vertical reactor body (1), at least one inlet (2) for the urea melt, a set of heating elements (3), and a central duct (7), **characterized in that**:

   said set of heating elements (3) is arranged inside said central duct.

2. Reactor according to claim 1, comprising an annular region (8) around said central duct, without heating elements.

3. Reactor according to claim 1 or 2, wherein said inlet (2) for the urea melt is connected to a distributor (10) configured to introduce urea into said annular region (8).

4. Reactor according to claim 3, said distributor (10) being configured to introduce urea in a substantially uniform manner and with axial symmetry into said annular region (8).

5. Reactor according to claim 4, wherein said distributor

(10) has a substantially toroidal form.

6. Reactor according to any one of the preceding claims, wherein said central duct (7) extends at a predefined distance from a base of the set of heating elements, thus defining a passage (9) for recirculating liquid between the central duct (7) and the surrounding annular region (8).

7. Reactor according to any one of the preceding claims, wherein said central duct (7) extends, inside the reactor, up to a height lower than the height of the heating elements (3).

8. Reactor according to any one of the preceding claims, wherein said central duct (7) is delimited by a substantially cylindrical shell (4).

9. Reactor according to any one of the preceding claims, wherein said set of heating elements (3) is a bundle of tubes connected to a tubesheet (5) at the bottom of the reactor.

10. Reactor according to any one of the preceding claims, **characterized in that**, during its functioning, the reactor has a descending flow of liquid inside the central duct (7) in the presence of the heating elements (3) and an ascending flow in the annular region (8) without heating elements around said central duct.

11. Reactor according to any one of the preceding claims, comprising a deflector (6), preferably flat or frustoconical, which is in the form of circular rim and is located at the top of said annular region (8).

12. Reactor according to claim 11, comprising a header for collecting the melamine (15), associated with said deflector and configured to collect melamine in a distributed manner along a circumference.

13. Process for the high-pressure synthesis of melamine from urea, comprising a primary reaction stage in which a stream of urea is converted into melamine with an endothermic reaction, inside a vertical reactor.
characterized in that:

said primary reaction stage is performed with a flow circulating inside a liquid mass comprising melamine and urea, said flow comprising:

a flow descending in a central portion (7) of said reactor, directly heated by the presence of heating elements (3) inside it, and
a flow ascending in a peripheral region (8) which is arranged around said central portion and is without heating elements and into which the urea supply (2) is introduced.

14. Process according to claim 13, wherein the pressure is at least 70 bar and preferably between 70 and 250 bar.

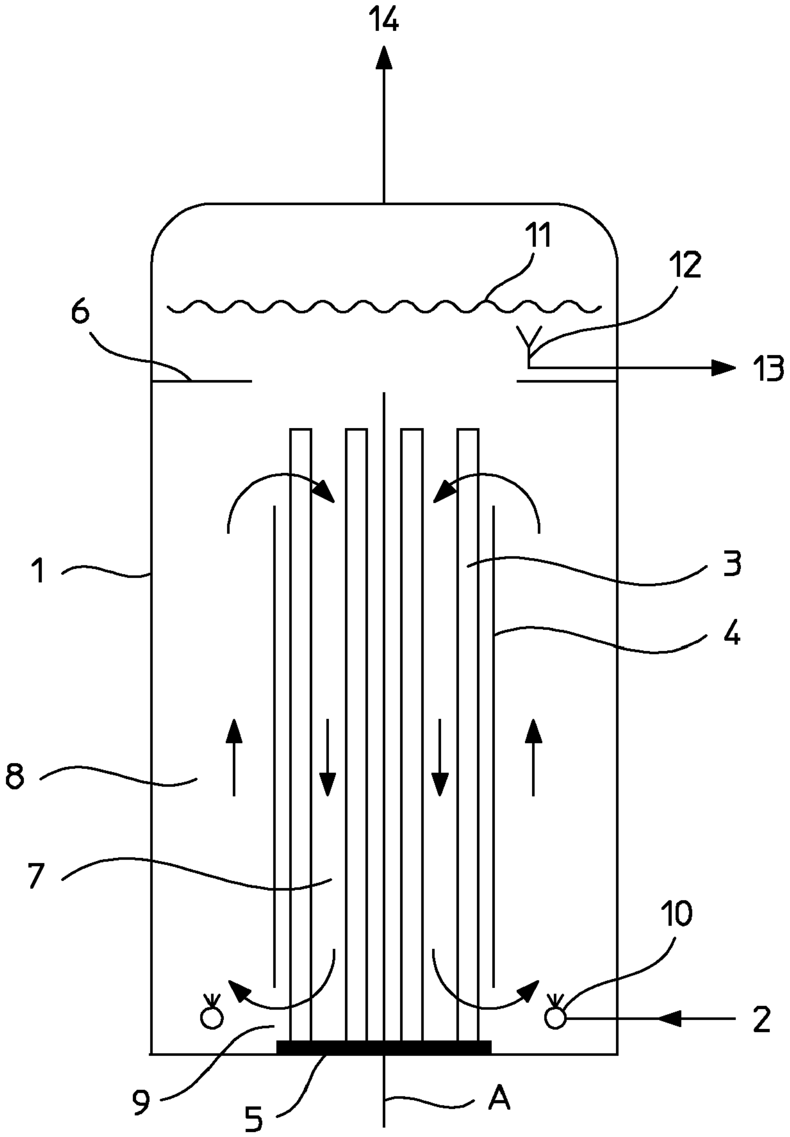

Fig.1

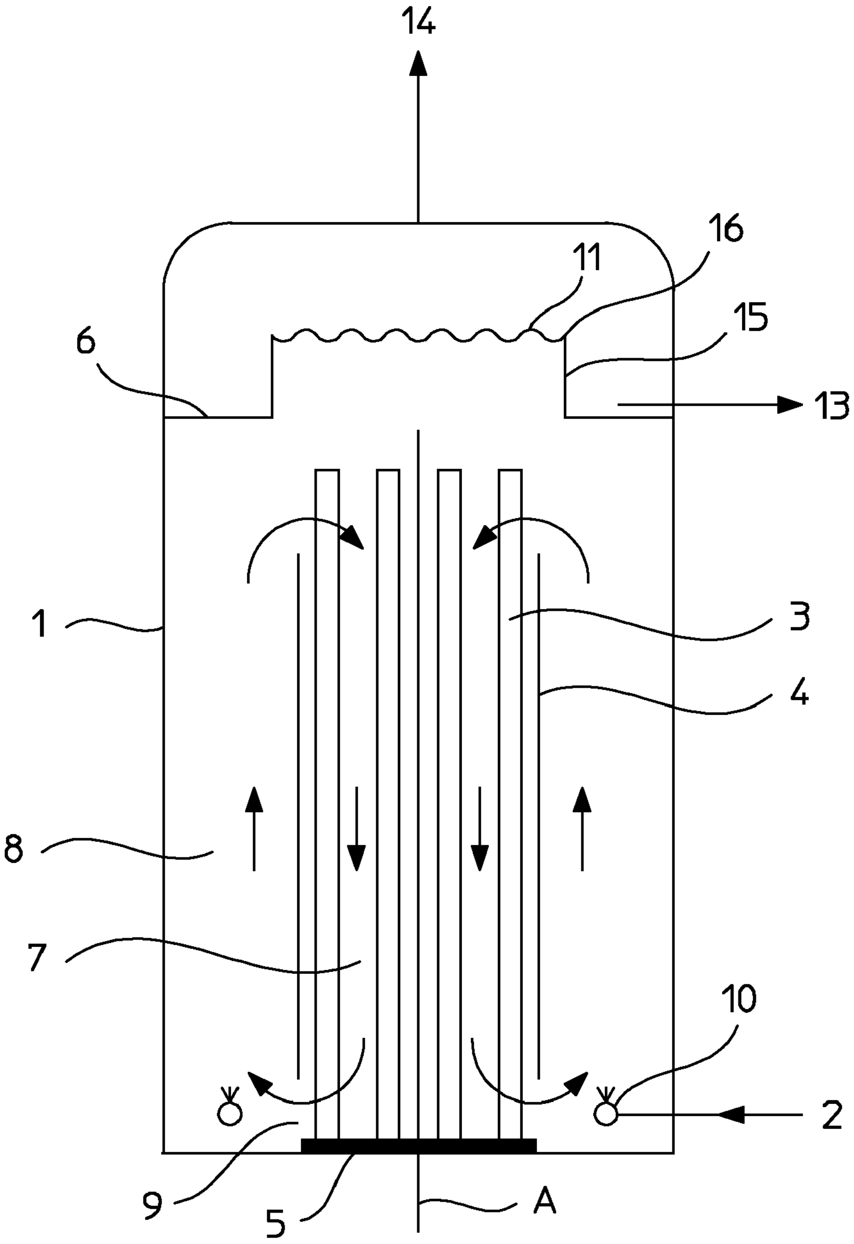

Fig.2

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EUROPEAN SEARCH REPORT

Application Number
EP 14 15 9283

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The present search report has been drawn up for all claims

1

Place of search	Date of completion of the search	Examiner
The Hague	4 July 2014	Baumlin, Sébastien

CATEGORY OF CITED DOCUMENTS

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Application Number
EP 14 15 9283

DOCUMENTS CONSIDERED TO BE RELEVANT

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Place of search	Date of completion of the search	Examiner
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